The Los Alamos Solid-State Optical Refrigerator

Richard I. Epstein, Bradley C. Edwards, Carl E. Mungan and Melvin I. Buchwald

Los Alamos National Laboratory
Los Alamos, New Mexico 87545

ABSTRACT

Optical refrigeration may provide the physical basis for solid-state cryocooling. Devices based on this physics would be vibrationless, compact, and free of electromagnetic interference. Having no moving parts and being pumped by a diode lasers, optical refrigerators would be rugged with operating lifetimes of years. Experiments at Los Alamos National Laboratory have demonstrated the basic physical principles of optical refrigeration.[1] Design studies suggest that optical refrigerators with efficiencies comparable to other small cryocoolers should be realizable with existing technologies.[2]

INTRODUCTION

The Los Alamos Solid-State Optical Refrigerator (LASSOR) concept has a laser diode pumping a *cooling element* which then radiates its heat through anti-Stokes fluorescence. That is, the cooling element absorbs photons at one wavelength and then emits photons at shorter wavelengths. The energy difference between the absorbed and emitted radiation is supplied by annihilating thermal phonons and thereby cooling the solid.[3,4] The complete refrigerator also requires some way of disposing of the fluorescence and a means of attaching the load to the cooling element.

One LASSOR design is described below, but many others can be imagined for specific applications.[5] Since optical refrigeration occurs on the atomic scale, it offers great flexibility in geometry and size. It should be possible to construct 10-100 watt cooling capacity LASSORs for cooling large mirrors as well as a milliwatt LASSORs for small electronic components. The optical refrigerators may even be integrated directly into the device to be cooled. For example, the glass of a mirror may be the LASSOR cooling element[6] or electronic components may be deposited directly on a cooling element.[7]

All LASSORs would be vibration-free. Their lifetimes would be limited by that of the diode laser (which can be many years[8]), and additional reliability can be achieved by using redundant lasers. The cooling efficiency, operating temperature and power density could vary over wide ranges depending on the atomic properties of the cooling element.

The remainder of this paper is organized as follows. We first discuss the basics of optical refrigeration and then describe the laboratory confirmation of these concepts. Finally, we present a design for a one-watt LASSOR device . Additional information on the LASSOR program can be found at the web site <http://sst.lanl.gov/~edwards/cooling.html>.

BASIC PRINCIPLES

The essentials of optical refrigeration. can be illustrated by the example of a three-level atom imbedded in an otherwise transparent solid (see Fig. 1). Laser light of photon energy $E_L = E_3 - E_2$ pumps an atom from energy level 2 to level 3. A subsequent radiative deexcitation moves the atom either to level 2 or to the ground state, level 1. In the former case the emitted photon has the same energy E_L as the absorbed laser photon and the system is unchanged. In the latter case the fluorescing photon carries away energy $E_F = E_3 - E_1 > E_L$ and there is a net shift of an excitation from level 2 to level 1. The relative populations of levels 1 and 2 are thus pushed out of thermal equilibrium. To restore thermal equilibrium the atom jumps from level 1 to 2 by absorbing a phonon of energy $E_P = E2 - E_1$, thereby decreasing the solid's thermal energy.

The three-level optical refrigerator cycle highlights the necessary and desirable properties of cooling materials. The foremost requirement is that the fluorescent efficiency be nearly unity. That is, an atom in level 3 must nearly always deexcite to level 1 or 2 by emitting a photon . The alternative is that the atom *nonradiatively* decays producing phonons whose total energy is much greater than $E_3 - E_2$ and can overwhelm the optical refrigeration effect. High fluorescent efficiencies are possible if the energy gap, $E_3 - E_2$, is large compared to the maximum phonon energy $E_{p,max}$ the solid can support. When this condition is satisfied, a nonradiative decay requires that at least $(E_3 - E_2)/E_{p,max}$ phonons are emitted simultaneously; such high-order processes are strongly inhibited. Additionally, it is vital that the transparent host be exceedingly pure. Small levels of certain contaminants can quench the fluorescence and lower the quantum efficiency.

The next requirement is that the populations of the lower energy levels remain close to thermal equilibrium through the emission and absorption of phonons. This is usually possible if the energy spacing $E_2 - E_1$ is not large compared to $E_{p,max}$. However, at low temperatures the phonon density falls off and the thermalization rate of the low lying levels can slow, limiting the performance of the optical refrigerator.

If the decay rates from level 3 to levels 1 and 2 are equal, then the average heat lift per pump photon is $0.5(E_2 - E_1)$, and the coefficient of performance of the refrigerator is

$$COP = 0.5\,(E_2 - E_1)/(E_3 - E_2)\,. \qquad (1)$$

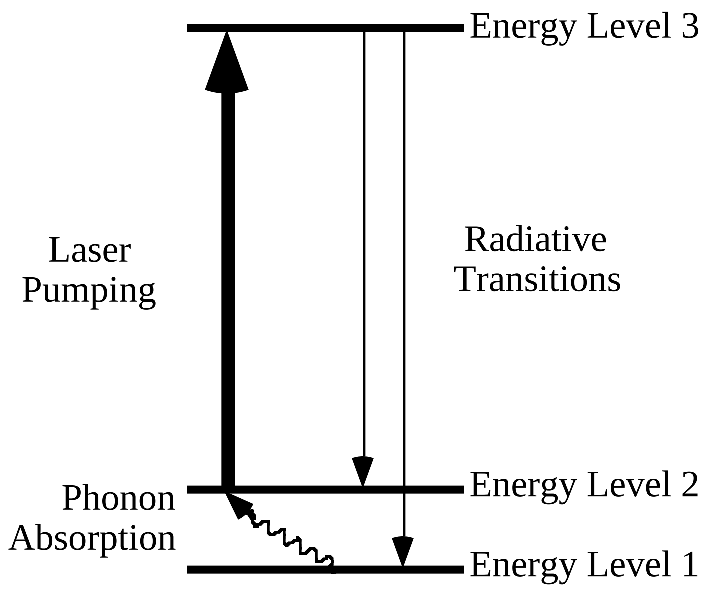

Figure 1. A three-level optical refrigerator

The refrigerator's efficiency is thus improved if the energy gap, E_3 - E_2, is decreased while the gap, E_2 - E_1, is increased. We see that there may be a trade off where the criteria for a high COP is in conflict with the conditions for a high fluorescent efficiency and rapid thermalization rate of the lower levels.

The optimal operating temperature of the refrigerator is in large part set by the size of the energy gap, E_2 - E_1. As the temperature decreases, the fraction of atoms in level 2 decreases approximately as the Boltzmann factor, $\exp[(E_1 - E_2)/kT]$, where k is the Boltzmann constant. Eventually, there would be too few atoms in level 2 to pump with the laser, and the refrigerator cannot operate. The minimum operating temperature would depend on the fluorescent efficiency, the atomic cross sections as well as the design of the cooler. As a rough guide, we expect the useful operating temperature Top to be approximately

$$T_{op} \sim 0.1(E_2 - E_1)/k . \qquad (2)$$

Comparing Eqs. (1) and (2) shows the tradeoff between efficiency and operating temperature, COP $\propto 1/T_{op}$.

LABORATORY STUDIES

In our experiments to date we use a heavy-metal-fluoride glass (ZBLAN) doped with trivalent ytterbium ions. Each Yb^{3+} ion possesses only two groups of energy levels below the UV absorption edge of the host glass.[9] These groups are separated by an energy of 1.3 eV corresponding to a wavelength of ~1000 nm. The ground-state group is split into four Stark levels and the excited-state group is split into three levels, as shown schematically in the inset of Fig. 2. The main part of this figure shows the measured absorption and fluorescence spectra of a sample of Yb doped ZBLAN. The mean energy of the fluorescent photons corresponds to a wavelength λ_F = 995 nm, as indicated by the vertical line. Pumping this glass in the long-wavelength tail of the absorption spectrum moves excitations from the top of the ground-state group to the bottom of the excited-state group. The relative populations of the Stark levels *within* each group are thereby shifted slightly out of thermal equilibrium. By absorbing phonons

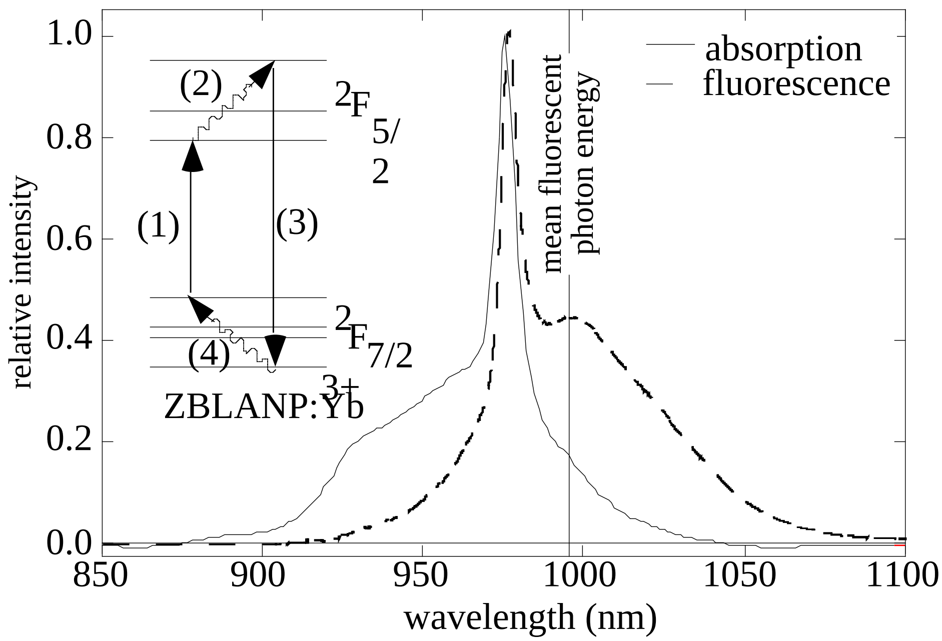

Figure 2. *Main plot*—Absorption coefficient (dashed curve) and fluorescence spectrum (solid curve) at room temperature for Yb doped ZBLAN glass The wavelength corresponding to the mean fluorescent-photon energy, λ_F, is indicated by a vertical line at 995nm. *Inset plot*—The schematic energy-level structure of Yb doped ZBLAN; the splittings within each group have been exaggerated for clarity. The arrows denote a typical cooling cycle: (1) laser pumping, (2) phonon absorption, (3) radiative decay, and (4) additional phonon absorption.

from the host material, thermal equilibrium within each group is restored. Radiative decays from the excited-state to the ground-state groups produce photons that carry off both the absorbed radiative and thermal energies.

Each fluorescent photon carries off, on average, thermal energy equal to the difference between the pump-photon and the mean fluorescent-photon energies. In the ideal case where there are no nonradiative relaxations from the excited- to the ground-state groups, the cooling power, P_{cool}, is proportional to the absorbed pump power, P_{abs}, and to the average difference in the photon energies of the pump and fluorescence radiation. In terms of wavelength λ of the pump radiation, the cooling power is

$$P_{\mathrm{cool}}(\lambda) = P_{\mathrm{abs}}(\lambda)\,(\lambda - \lambda_F)\,/\,\lambda_F\,. \tag{3}$$

Two separate experimental arrangements were used to test the validity of the optical refrigeration relation, Eq. (3). The specifics of these two experiments can be found in ref. 1. The main results of these experiments is shown in Fig. 3. which shows the cooling efficiency, defined as the ratio of the cooling power to the absorbed laser power, that was measured with the two experiments. In the first of these experiments, photothermal-deflection spectroscopy[10] was employed to measure the temperature changes induced by the pump laser in the interior of a sample. The pump beam from a ~ 1 micron cw laser was focused into the sample and heated or cooled a thin cylindrical column. The temperature change produced a small density change in the illuminated region thereby generating a weak lens. A helium-neon laser probe beam was directed through the sample so that it could be deflected by the thermally induced lens. Angular deflections of this probe beam were measured with a position-sensitive photodetector. An optical chopper placed in the path of the pump beam modulated the photothermal-deflection signal. The direction of the deflection indicated whether the sample was heating or cooling, and the amplitude of the deflection was proportional to the magnitude of the temperature change.

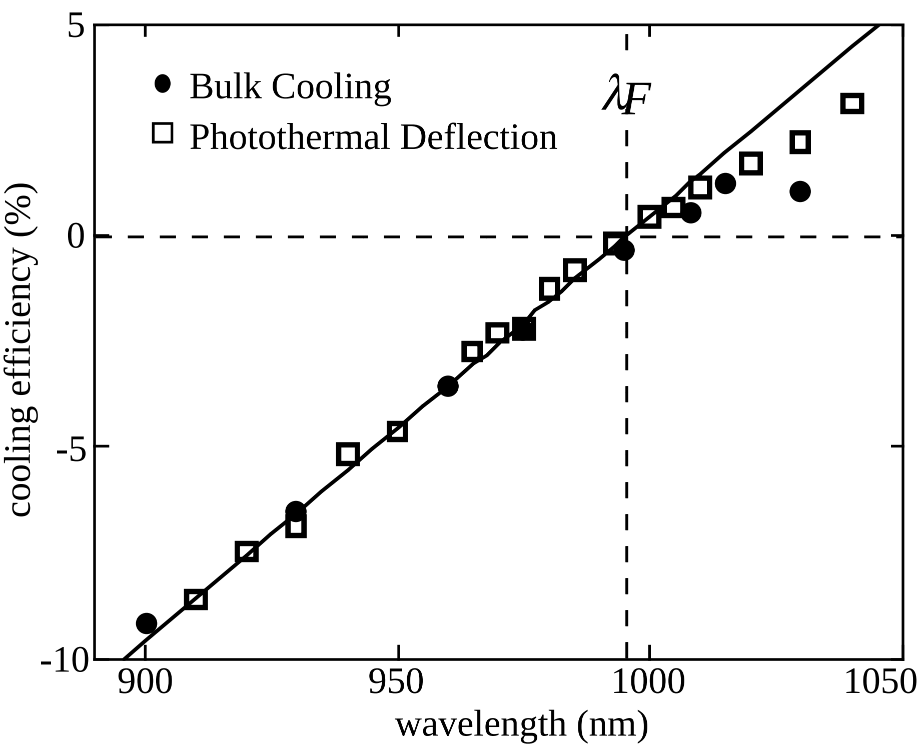

Figure 3. Cooling efficiencies measured in the photothermal-deflection (filled circles) and bulk-cooling (open squares) experiments. Negative efficiencies correspond to heating. The solid curve is a plot of Pcool/Pabs from Eq. (3).

In the second experimental arrangement, the equilibrium temperatures of a bulk $2.5 \times 2.5 \times 6.9\ mm^3$ sample were measured when the sample was continuously pumped by a laser beam. The sample was positioned at the center of a vacuum chamber and rested on two thin, vertical glass slides that limited the conductive heat transport from the chamber walls. The inner wall of the chamber was painted black to absorb stray pump radiation as well as the fluorescence emitted by the sample. The pump-laser beam is directed along the long axis of the sample. The temperature of the sample was monitored with an infrared camera. To account for the changes in the sample temperature arising from temperature drifts of the chamber, a reference sample was positioned ~1 cm away from the test sample, outside the pump-beam path but on the same glass supports in the chamber. Temperature differences of 0.02 K between the pumped and reference samples were resolvable.

The equilibrium temperature, T_S, of the sample is established by a balance between the optical refrigeration and the heat load from the environment. In our setup, the dominant heat load was from the radiative coupling between the walls of the vacuum chamber at ambient temperature T_C and the sample. If the sample and walls radiate as blackbodies, then

$$P_{\text{cool}} = \sigma A(T_C^4 - T_S^4), \tag{4}$$

where σ is the Stefan-Boltzmann constant and A is the surface area of the sample.

Fig 3. shows that the results of the photothermal deflection and bulk cooling experiments were in good agreement with each other and with the predictions of Eq. (3). In agreement with the theory, most of the data lie on a straight line. If the quantum efficiency were unity, the zero crossing would occur at λ_F = 995 nm. Experimentally, the zero crossing agrees with this prediction to within 3 nm, indicating that the fluorescence quantum efficiency is at least 0.997. The deviations from linearity for $\lambda \geq$ 1020 nm may be due to parasitic heating, perhaps from surface contamination.

Our most recent photothermal deflection experiments, which were carried out at cryogenic temperatures, have shown cooling efficiencies comparable to those of Fig. 3. Additionally, recent bulk cooling experiments using ZBLAN fibers have shown temperature drops more than a factor of 40 greater than those discussed in ref. 1. The specifics of these experiments will be reported elsewhere.

LASSOR DESIGN

Since a solid-state refrigerator is particularly well suited for space applications, our initial designs have focused on these applications. Figure 4 illustrates a 1-watt version of the Los Alamos Solid-State Optical Refrigerator (LASSOR) that might be used for cooling a space-borne infrared detector or a germanium gamma-ray spectrometer to 77K.

This design is based on ytterbium doped ZBLAN glass. A diode laser coupled with an optical fiber delivers ~1020 nm pump radiation to the cooling element. The fiber carrying the pump radiation passes through the wall of the cryocooler and to a cylindrical block of Yb doped ZBLAN. Dielectric mirrors that reflect the pump radiation are deposited on both ends of the cooling element. The pump radiation enters the cooling element through a small hole in one of the mirrors. This radiation is directed parallel to the axis of the cylinder so that it is repeatedly reflected from the dielectric mirrors and from the sides of the glass cylinder (by total internal reflection). Ultimately, the pump radiation is absorbed by Yb ions, and the doped glass cools as it fluoresces. Most of the isotropic fluorescent radiation escapes from the cooling element and is absorbed by the warm walls of the cooling chamber. A metal mirror is attached on the back of the upper dielectric mirror creating a completely shadowed region where the cold finger can be mounted. The inner surfaces of the chamber walls contain a coating that readily absorbs 1 micron radiation while having a very

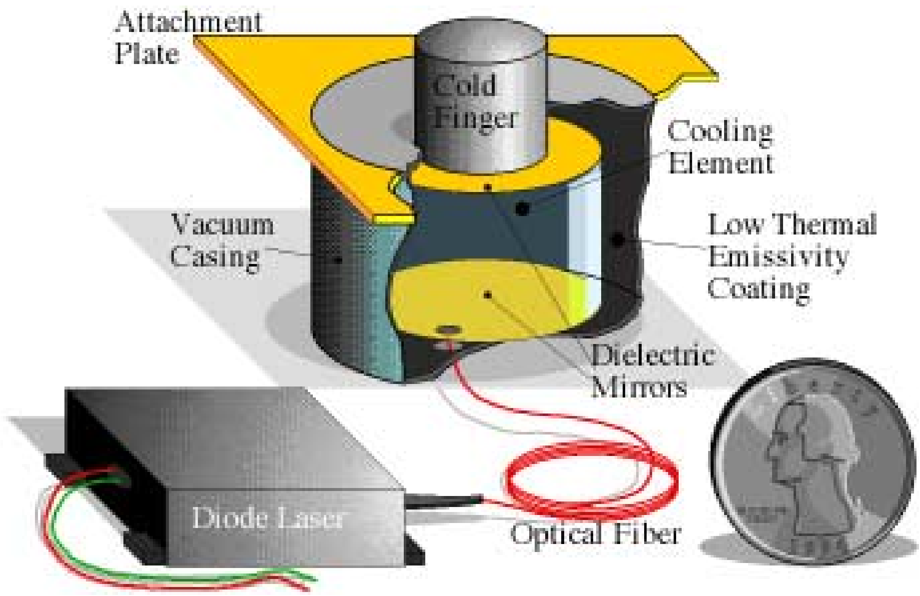

Figure 4. A design for a 1-watt LASSOR

high reflectivity for ~10 micron thermal emission. This coating would lessen the radiative heat load from the warm chamber walls.

We estimate that a practical, first-generation, optical refrigerator of the type described here could operate near 77 K with almost 0.5% efficiency (DC power to cooling power), weigh less than 2 kg/W and have a lifetime of 10 years of continuous operation.[2]

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